

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
 - TEXT CUT OFF AT TOP, BOTTOM OR SIDES
 - FADED TEXT
 - ILLEGIBLE TEXT
 - SKEWED/SLANTED IMAGES
 - COLORED PHOTOS
 - BLACK OR VERY BLACK AND WHITE DARK PHOTOS
 - GRAY SCALE DOCUMENTS
-

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problems Mailbox.**

THIS PAGE BLANK (USPTO)

PRVPATENT- OCH REGISTRERINGSVERKET
Patentavdelningen

SE 99/01084

S

**Intyg
Certificate**

| | |
|-------------------|-----|
| REC'D 03 AUG 1999 | |
| WIPO | PCT |



Härmed intygas att bifogade kopior överensstämmer med de handlingar som ursprungligen ingivits till Patent- och registreringsverket i nedannämnda ansökan.

This is to certify that the annexed is a true copy of the documents as originally filed with the Patent- and Registration Office in connection with the following patent application.

(71) Sökande Akzo Nobel NV, Arnhem NL
Applicant (s)

(21) Patentansökningsnummer 9802224-7
Patent application number

(86) Ingivningsdatum 1998-06-22
Date of filing

Stockholm, 1999-05-14

För Patent- och registreringsverket.
For the Patent- and Registration Office

Evy Morin
Evy Morin

Avgift
Fee 170:-

**PRIORITY
DOCUMENT**
SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH RULE 17.1(a) OR (b)

Ink. t. Patent- och reg.verket

Akzo Nobel NV

1998-06-22

Method for application of a gluing system

Huvudförfattaren Kossan /

The present invention relates to a method for separate application of the components of an expandable gluing system onto substrates, the hardener of which system is acidic and forms part of the expandable capability of the system.

Prior art

Expandable gluing systems are known in the art. Such expandable gluing systems can for example be used where pieces to be glued together exhibit irregularities in the surfaces thereof to which glue is to be applied, such as for example in the gluing of wooden pieces together, such as, for instance, for the manufacture of gluelam or laminated timber. The glue will then fill out these irregularities and thereby provide a joint having an improved strength as compared to a gluing system without a gas-forming substance. Another object of such systems is to reduce the amount of wood which has to be removed by means of, for instance, planing before gluing in order to make the surfaces to be glued smooth. However, in known expandable gluing systems the expandable feature is accomplished by means of, for example, a gas-forming composition or mixture separate from the resin and hardener components, respectively, which is introduced into the gluing system at the time of gluing. Such an expandable gluing system, more particularly a carbamide resin based system, has been disclosed in SU-327224, wherein the mixing of a foaming composition with the resin suitably can be effected at the time of introduction of the carbamide resin curing catalyst.

Hitherto, expandable gluing systems has only been applied to substrates in the form of an admixture of the components included in said system. The expandable systems known in the art, will, for example, be difficult to recirculate, due to, inter alia, the risk of clogging, or build up in the application apparatus of hardened, or partially hardened,

1998-06-22

Huvudfaxen Kassa /

mixture of the components of the gluing system. Additionally, the risk of foaming, or undesired gas formation in the application equipment will be present due to the presence of the foaming composition. Also start and stop of such application, and operation during long periods of time, will be difficult in practise.

Accordingly, it would be highly desirable to provide a method for application of an expandable gluing system, wherein the components of said system can be recirculated, and wherein the formation of expanding agent, i.e. gas, prior to the pressing of the glued substrates together, is minimized. Also, it would be highly desirable to be able to uniformly regulate the process of expansion, i.e. gas formation, for example, in order to allow relatively long periods of time from application to pressing, and to secure a uniform formation of gas in the applied components, especially during the pressing.

Thus, according to the present invention, a method for separate application of the components of an expandable gluing system, comprising a resin component and an acidic hardener component is provided, wherein the resin component comprises a gas generating substance, which substance is capable of forming a gaseous expanding agent, such as, for example, carbon dioxide, when contacted with the acidic hardener component of the system. The acidic hardener component used in the method of the invention comprises an acid. An especially suitable group of acids are the organic acids. Suitable examples of the latter are paratoluene sulfonic acid and carboxylic acids, the latter of which being preferred. Examples of suitable organic carboxylic acids are, formic acid, citric acid, and maleic acid, of which formic acid, and maleic acid are preferred.

In this manner, the hardener used also has the additional function of providing a component of the reaction system generating the gas, the other component of the gas-producing reaction system being the gas generating substance. Said gas generating substance is provided in admixture with the resin component of the gluing system. The present invention thus,

inter alia, offers the advantages of using an expandable gluing system having a reduced number of essential constituents. Additionally, according to the invention, the need of a step comprising the addition of a gas-forming system into the gluing system, or into any of its components, prior to application of the gluing system, can conveniently be eliminated.

The technique according to the present invention is especially useful with the amino resin systems, the hardeners of which are generally acidic. Examples of such systems are; urea-formaldehyde, melamine-urea-formaldehyde, melamine-formaldehyde, melamine-urea-phenol-formaldehyde resins, and furfuryl alcohol modified varieties thereof.

The gas producing substance can be any carbonate or hydrogen carbonate that will generate CO₂ on contact with an acid. Suitable examples are, for instance, sodium hydrogen carbonate, calcium carbonate, sodium carbonate, ammonium carbonate, ammonium hydrogen carbonate, magnesium carbonate, or a mixture thereof. Preferably calcium and/or sodium carbonate is used. The gas producing substance can suitably be included in the resin component in an amount ranging from 0.1-10 % by weight, as calculated on the pure, active form of said substance, ie. the form that will generate the gas, with any impurities excluded.

Other gas generating substances, which generate a gas having a low boiling point on the contact with an acid used in the hardener, could also be used in the present invention. Examples of such gases are, for instance, ammonia, nitric dioxide, etc, although not preferred from an environmental standpoint, for example.

The gas generating substance can also, for example, be used together with a conventional filler, or mixtures thereof. However, when the filler used comprises, or constitutes, one or more gas generating substances as used herein, the total amount of the pure active form of such substances should be kept within the interval stated above.

1998-06-22

Huvudfaxen Kassan /

In the method of the invention, the two components constituting the gluing system are applied separately. Application of the gluing system can be accomplished by means of curtain, spray or strand type of application, or any combination thereof. Although possible, the use of rollers is not preferred, mainly due to the risk of one of the components getting in contact with the other on one of the rollers. Also, the application speed would be limited by the risk of misting or flying occurring at the rollers. Application in the form of separate strands of the respective components is preferred.

As used herein, the term "strand" also comprehends the meaning of the term "ribbon", also conventionally used in the art, and any other like term.

After application of the expandable gluing system to the substrates, such as, for example, wooden lamellae, said substrates are brought together and pressed into an aggregate, such as, for example, a wooden beam.

It is preferred that the main production of gas in the expandable gluing system essentially occurs at the time of pressing, following the application of said system to the substrates. Since the gas-forming reaction commences upon contact of the two components, the contact of said components with each other should desirably not be fully established until such pressing. Thus, the method of the invention employs separate application in order to minimize the contact and the degree of mixing of the components prior to pressing, and thereby the extent of the gas-forming process occurring prior to pressing. Preferably, a regulated, continuous, minimal contact of the different strands should be secured, in order to secure an adequate miscibility of the strands during pressing. Thereby, the gas-forming reaction can be more carefully regulated, while also longer periods of time are allowed to lapse between application and pressing, such as, up to 180 minutes, if desired, preferably 0-90 minutes. This object is

1998-06-22

5

Huvudföran Kassan

achieved by using application of the components in the form of strands.

During the pressing, the two components will flow to some extent between the pieces of substrate that are being pressed together, whereby the components also will be mixed with each other to a higher degree than prior to the pressing, and are thereby brought into a more intimate contact with each other. In this manner the principal gas-generation will occur during the pressing. Also, the gas formation during the pressing will contribute to the mixing to some extent.

Accordingly, in one embodiment of the method of the present invention, separate application of the components in the form of strands is used. A suitable device for such application of the components used in the present method, is a device comprising a unit of at least two hollow members, at least one member for each component, provided with a number of orifices, from which orifices in each member the respective component is applied to a substrate below said hollow members, wherein the hollow members are connected to each other by fixation means, said members being positioned above the plane of application, wherein each of the holes in one of the members are aligned in the machine direction with the corresponding holes in the other member(s).

A preferred method of separate application in the form of strands is a method wherein the above device is used, whereby the corresponding strands of the components used, are in essentially continuous contact with each other throughout the length of said strands.

In this manner, uniform contact of the resin and hardener strands is secured at the time of application, while the more fully intermixing of the two components, generating the gas, is not established until the time of pressing.

As an example of another suitable embodiment of the method of the invention, a method wherein the resin component preferably

Huvudföran Kassar
is applied first, in the form of strands, which strands optionally can coalesce to form an essentially continuous layer thereof, whereafter the hardener component, in the form of strands, or by means of spraying, is applied, can be used. This embodiment is, for example, suitable when it is desired to minimize the contact of hardener and substrate.

In the following two examples, roughly planed pieces of spruce were provided with a conventional non-expandable gluing system, and with an expandable system of the invention, respectively, by means of separate strand application of the two components after each other. Thereafter laminates were formed from the pieces with the non-expandable, and the expandable gluing system, respectively, and subsequently tested for delamination.

Example 1

Substrate:

90 cm x 15,5 cm pieces of spruce

Resin component:

SL97044 (a melamine-urea formaldehyde resin, containing calcium carbonate as the gas generating substance)

Content of gas
generating substance

in the resin:

1 % by weight

Hardener component:

formic acid based

Molar ratio of resin/hardener

100:30

Amount applied:

400 g/m²

Order of application:

resin, then hardener

After application, the laminates were pressed at a pressure of 7-8 bar during the night. After one day of after-curing the laminates were tested for delamination. The results are shown in the Table below.

Example 2 (comparative)

Example 1 was repeated with the only difference that the resin component used was SL97043 (a melamine-urea-formaldehyde resin

1998-06-22

containing no gas generating substance). The resulting laminates were tested in the same way as described above, and the results are given in the Table below.

| Gluing system=SL 97043/formic acid based hardener | | Gluing system=SL97044*/formic acid based hardener (expandable) |
|---|-------|--|
| Laminate=A | 18,3% | 3,9% |
| B | 3,21% | 0,7% |
| C | 0% | 0,7 |
| D | 0,5% | 0 |
| E | 2,9% | 0 |
| F | 3,4% | 1,3 |
| G | 6,3% | 1,3 |
| H | 5,6% | 0 |
| I | 7,5% | 0,5 |
| Average: | 5,9% | 0,9% |

* SL97044 = SL97043 + gas generating substance

From the table, it can be clearly seen that the delamination results for the laminates formed using the gluing system according to the present invention are substantially improved, as compared to those for the laminates formed with the non-expandable gluing system. The delamination was determined according to EN-391-B.

Thus, these examples clearly demonstrate the improvements obtained with the method of the present invention, wherein an expandable gluing system is used, as compared to the usage of a non-expandable gluing system.

1998 -06- 24

Claims

Huvudföresättnen Kassen

1. Method of gluing pieces of substrate together, comprising separate application of resin and hardener components of an expandable gluing system, characterized in that said hardener component is acidic, and the resin component comprises one or more gas generating substances capable of forming a gas when contacted with said hardener component.
2. Method according to claim 1, characterized in that the gas generating substance(s) is included in the resin component in an amount in the interval of 0.1-10 % by weight, as calculated on the pure, active form of said substance(s).
3. Method according to claim 1 or 2, characterized in that the gas generating substance(s) is a carbonate or hydrogen carbonate which generates CO₂ on contact with an acid.
4. Method according to claim 3, characterized in that the gas generating substance is calcium and/or sodium carbonate.
5. Method according to any of the previous claims, characterized in that the gluing system used is an amino resin gluing system.
6. Method according to claim 5, characterized in that the amino resin gluing system is selected from the group of melamine-urea-formaldehyde, melamine-formaldehyde, and urea-formaldehyde resin gluing systems.
7. Method according to claim 5 or 6, characterized in that the resin component is applied in the form of strands, and the hardener component is applied in the form of strands, or by means of spraying, in optional order of application, wherein the first applied component optionally can coalesce to form an essentially continuous layer thereof, onto which the other component is applied.

Ink. t. Patent- och reg.verket

9

1998-06-22

Huvudfören Kassan /

8. Method according to any of the claims 1-6, c h a r a c t e r i z e d in that both components are separately applied in the form of strands by means of a device comprising a unit of at least two hollow members, at least one member for each component, provided with a number of orifices, from which orifices in each member the respective component is applied to a substrate below said hollow members, wherein the hollow members are connected to each other, said members being positioned above the plane of application, wherein each of the holes in one of the members are aligned in the machine direction with the corresponding holes in the other member(s).

9. Method according to any of the previous claims, c h a r a c t e r i z e d in that the substrates are made of wood.

10. Method according to any of the previous claims, c h a r a c t e r i z e d in that the main formation of the expanding agent occurs during pressing of the pieces, to which said gluing system components are applied, for gluing them together, in order to prepare a glued aggregate.

'98 08/22 16:41

46 8 4291070

ARRANGENTBYRÅ PATENTVERKET

013

46 8 4291070

10

Ink. t. Patent- och reg.verket

1998-06-22

Huvudföres. Kassen

ABSTRACT

A method for separate application of the components of an expandable gluing system onto substrates is disclosed, the hardener of which system is acidic and forms part of the expandable capability of the system.

THIS PAGE BLANK (USPTO)